Scattering of low-energy helium atoms from a low-temperature solid surface

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The first measurements are presented of the scattering of low-energy ($\lesssim 2$ meV) He atoms from a solid surface at low temperature ($T_s = 3.6$ K). The results are consistent with calculations which assume that all incident atoms either stick with probability α or are scattered elastically and specularly. As the mean incident angle with respect to the surface normal decreases from 45° to 14°, α increases from 0.77 to 0.94.

The scattering of ^{4}He atoms from solid surfaces is of considerable interest as a probe of surface static and dynamical properties.[1,2] One regime which is particularly valuable is that of low incident energy and surface temperature T_s. The reason is that multiphonon and diffraction effects are expected to play a relatively small role in these circumstances. Unfortunately, cryogenics complicates the already difficult technique of producing and scattering supersonic molecular beams in high vacuum. Thus the important regime of low velocity and T_s are relatively unexplored by this otherwise powerful technique.

For the last few years, our group has been studying He desorption from surfaces.[3,4] We have recently extended our attention to the problem of He incidence on surfaces. An intriguing discovery of high sticking probability ($\frac{2}{3} < \alpha < 1$) for a thermal distribution of atoms ($10 < T_{\rm inc} < 20$ K) incident on a low-temperature surface ($T_s < 4$ K) has already been reported.[5] This paper presents the first results of a complimentary study of the *scattering* of such a ^{4}He beam. The data are consistent with primarily elastic and specular scattering of the (10–20%) atoms which do not stick.

The technique shown in Fig. 1 (inset) is an adaptation of that used in the desorption and sticking experiments. In all of these, the surfaces are optically flat, but are not otherwise characterized. A He film of approximately one monolayer or less covers all surfaces.[4] A short (30-ns) voltage pulse is applied to a fast ($\sim$10-ns) heater ($\sim$600-Å nichrome film) deposited on a sapphire crystal. The desorbing He atoms ballistically[4] traverse a distance l to the upper surface. The flux of those atoms which stick is monitored by a superconducting transition bolometer b_1 (0.3 mm square), the center of which is displaced 0.3 mm from the vertical axis of the heater. The reflected atom's flux is proportional to the signal S_2 at the bolometer b_2, which is laterally displaced 0.6 mm from the heater. The thermal time constant of b_1 and b_2 is sufficiently short ($\sim$10 ns), so that their response is proportional to the instantaneous flow. The sensitivities of the two bolometers coincide within 10%. Figure 1 shows the signal detected at b_1 and b_2 as a function of time after a 10.7-K heat pulse is applied to h. The signal at b_1 is due to atoms that stick there, and the signal at b_2 is due to atoms reflected from the upper surface.

We have computed the signals at b_1 and b_2 on the basis of three different models of the fate of those atoms that do not stick at the upper surface.

(1) They are reflected elastically and specularly.
(2) They are scattered elastically but diffusely.
(3) They are instantaneously redesorbed stochastically, with energies given by the beam temperature.

In performing these calculations we have assumed that the number of atoms desorbing from h with velocity in d^3v is

$$dN(\vec{v}) = Cv_z e^{-\beta E} f(\theta) d^3v \ , \tag{1}$$

where E is the kinetic energy, θ is the angle relative to the surface normal, β^{-1} is Boltzmann's constant times a temperature characterizing the desorption, $T_d = 10.7$ K in these experiments, and C is a normalization constant. The angle dependence is taken to be $f(\theta) = \exp(-\gamma \tan^2\theta)$, a reasonable form chosen for mathematical convenience. Equation (1) represents the results of our previous studies of desorption, giving a Maxwellian distribution focused normal to the surface.[3–7] The value of the parameter $\gamma = 1.06$ was derived from the data at $T_d = 10.7$ K taken in a separate desorption experiment.[6] The signals $S_1(t)$ and $S_2(t)$ at b_1 and b_2 are

$$S_1(t) = \alpha(E_z + E_b)\left[\frac{dN(t)}{dt}\right]_1 , \tag{2a}$$

$$S_2(t) = \alpha(1-\alpha)(E_z + E_b)\left[\frac{dN(t)}{dt}\right]_2 , \tag{2b}$$

where $dN(t)/dt$ is the rate of atoms arriving at the bolometers, derived from Eq. (1), integrated over the heater and bolometer areas. The term E_z is the part of the atom kinetic energy due to motion along the z direction, E_b is its binding energy to the surface, and α is the sticking coefficient.

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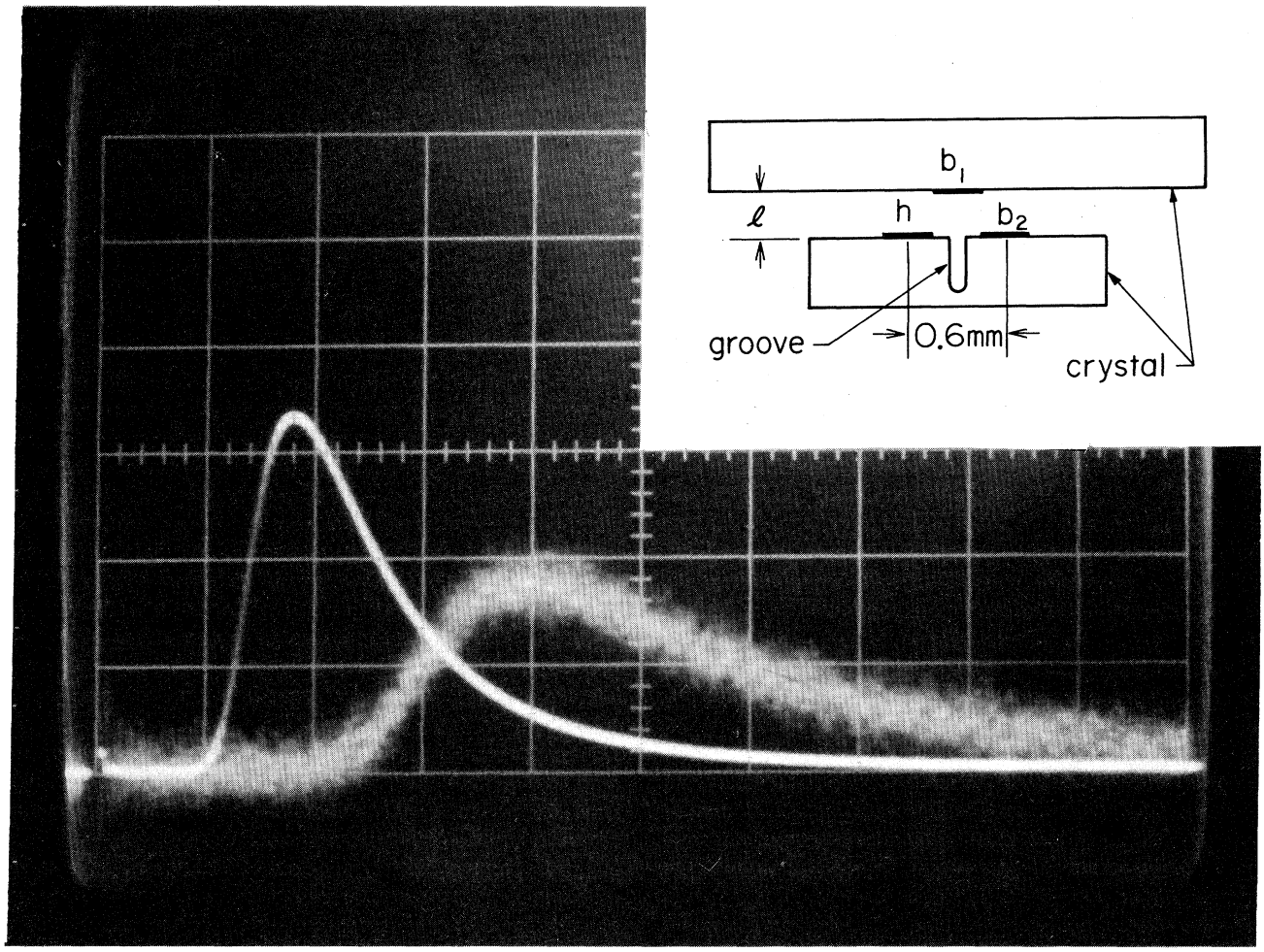

FIG. 1. Signals S_1 and S_2 (arbitrary units) measured by bolometers b_1 and b_2 as a function of time (1 μs per division) after heater-pulse-induced desorption; there is a factor of 25 greater relative amplification of S_2. Data shown are for vertical separation $l=0.6$ mm. Inset shows experimental design.

In Figure 2, the data for $l=0.6$ mm are compared with the computed signals. The maximum heights of signals S_1 and S_2 determine C and α in Eqs. (1) and (2), but the time of flight (e.g., the position of the maximum) of S_1 is determined by Eq. (2a), and that of S_2 is determined by the scattering model used. The elastic specular model, which gives the shortest arrival times, fits the data best. The elastic diffuse model predicts distinctly later times (see Fig. 2), and the stochastic model (not shown) gives still later times. Both alternative models predict broader distributions than those actually observed.

The data at $l=0.3$ mm are shown in Fig. 3. The heights of S_1 and S_2 are computed using the same value of C as in Fig. 2, but with a new value of α corresponding to a new angle of incidence. Thus a single parameter gives the absolute height of both curves. Computation of the times of flight in this geometry is complicated by the fact that the path from h to b_1 is at a sufficiently large angle θ to have a substantial "rainbow effect,"[7] i.e., the atoms emitted at this angle have a subthermal energy distribution easily detected in the data. Although difficult to account for analytically, this effect is easily eliminated from the scattering analysis by plotting the time in units of

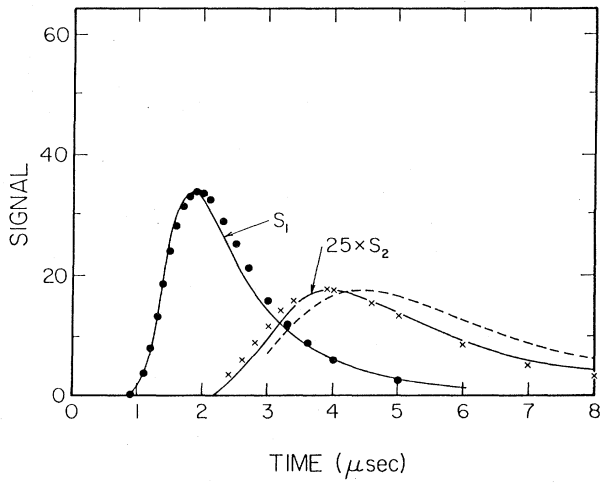

FIG. 2. Experimental signals (solid curves) for $l=0.6$ mm compared to signals calculated with elastic specular model with $\alpha=0.827$ and $T_d=10.7$ K (pluses). The dashed curve is calculated with the diffuse scattering model, normalized to agree with the measured S_2. Only one scale factor has been used.

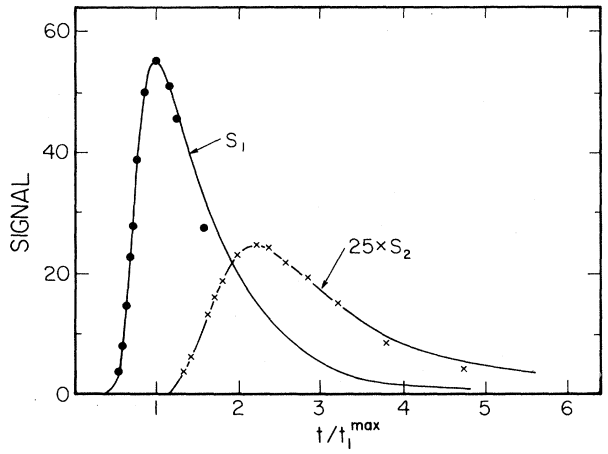

FIG 3. Signals at $l=0.3$ mm vs $t/t_1^{\max}$ with the same scale factor as in Fig. 2 and $\alpha=0.774$.

$t_1^{\max}$, the (somewhat delayed) arrival time of the maximum in S_1. When this is done the result once again is in excellent agreement with the elastic specular model.

In addition to the data shown in Figs. 1–3, measurements were made at $l=1.25$ mm. In this case, the reflected signal was so small that an accurate time of flight could not be determined, and the maximum height had an uncertainty of roughly $\pm 20\%$. Fortunately, however, it was still possible to deduce an accurate value of α from these data.

In practice, for each value of l, and hence of mean incident angle $\bar{\theta}$, the value of α is deduced from the ratio of maximum bolometer signals, S_2/S_1. Proceeding on the assumption of the elastic specular model, we have

$$S_2/S_1=(1-\alpha)F_0(l) \ . \tag{3}$$

$F_0(l)$ is determined by solid angle effects, the finite size of source and detectors, the focusing of the desorbed beam, and the spreading out of the signal in time due to the Maxwellian distribution of velocities of the desorbed atoms. We have computed F_0 using Eq. (2) and including these effects.[8,9] The results of the calculation are $F_0(l)=0.0803$, 0.126, and 0.128 for l equal to 0.3, 0.6, and 1.25 mm, respectively. These values may be compared to the value of 0.125 that would be expected for point source and detector.

TABLE I. Calculated $F_0(l)$, observed ratio S_2/S_1, and sticking coefficient α deduced from Eq. (3) for various l. The mean incident angle is $\bar{\theta}$.

l (mm)	F_0	S_2/S_1	α	$\bar{\theta}$ (deg)
0.3	0.0803	0.0182	0.774	45
0.6	0.126	0.0127	0.827	26.6
1.25	0.128	0.0083	0.935	13.5

The final results of this analysis are listed in Table I. For each l we show the measured ratio S_2/S_1, F_0, the value of α deduced from Eq. (2), and the mean angle with respect to the surface normal of the assumed specular reflection. We find that α increases from ~ 0.77 to ~ 0.94 as $\bar{\theta}$ decreases from 45° to 14°.

In summary, we find that helium atoms with kinetic energies centered at ~ 11 K incident on a surface at 3.6 K stick with high probability, but those that do not stick are scattered specularly and elastically, or nearly so. The sticking probability increases toward unity as the incident angle approaches the surface normal. Qualitatively similar behavior has been found in ^{4}He scattering from a liquid-^{4}He surface.[10,11] However, aside from the fact that the surface was a liquid, that experiment differed from ours in that it used even lower temperatures and found even higher α at all angles.

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[6]This corresponds to a $(\cos\theta)^{3.75}$ dependence of the desorption rate in Eq. (1). The separate experiment was done at $l=1.25$ mm where the heater and the bolometer are pointlike.

[7]Data of P. Taborek [Phys. Rev. Lett. 48, 1737 (1982)] at higher $T_d\sim 14$ K are fit by a value $\gamma\simeq 8$. The ambient temperature was lower (2 K) in that experiment. The difference in γ reflects the nonequilibrium character of the desorption. See Ref. 4.

[8]Details of these calculations will be presented elsewhere along with a more complete description of the experiments.

[9]The desorption from h has been assumed to be instantaneous. This is based on data of Ref. 3, in which even the very short (30-ns) pulse-width data conform to desorption at the heater temperature. See D. L. Goodstein and M. Weimer, Surf. Sci. 125, 227 (1983) for an explanation of this behavior.

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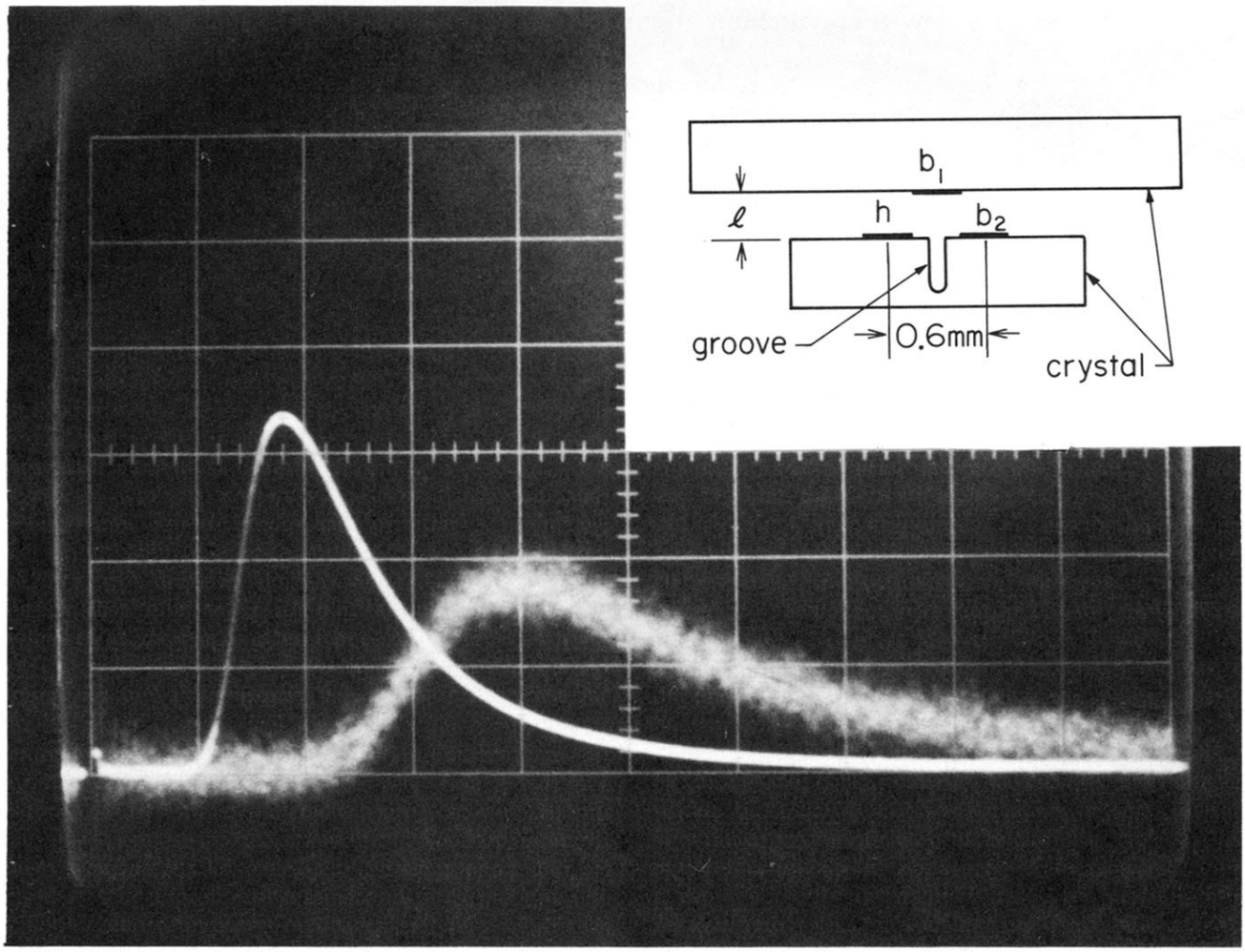

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